

MICROFILTRATION HYBRID SYSTEMS IN WASTEWATER TREATMENT FOR REUSE

Wenshan Guo

**Submitted in fulfillment for the degree of
Doctor of Philosophy**



University of Technology, Sydney

Faculty of Engineering

Australia

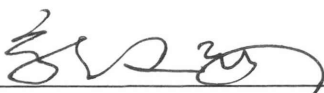
2005

CERTIFICATE OF AUTHORSHIP/ORIGINALITY

I certify that the work in this thesis has not previously been submitted for a degree nor has it been submitted as part of requirements for a degree except as fully acknowledged within the text.

I also certify that the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

Signature of Candidate



A handwritten signature in black ink, appearing to be 'S. V. 20', is written over a horizontal line. A long, sweeping flourish extends from the end of the signature down and to the left.

I dedicate this work to my parents

(Deyi Fu and Liren Guo)

&

Youhao Wu



ACKNOWLEDGEMENTS

I would like express my profound gratitude and appreciation to my principal supervisor, Professor Saravanamuthu Vigneswaran, for his valuable suggestions, ideas, supports, comments and continuous guidance throughout my study. I am very sincerely grateful to my other supervisor, Dr. Huu-Hao Ngo. It is very kind of him to give me so much supports, creative ideas, invaluable advices, enlightenment and encouragement. This work would not have been done without them.

The experimental work of this study was made possible through the funding of Australian Research Council (ARC) Discovery Grant (no. DP0211955, 2002–2003) and through the MOU of the University of Technology, Sydney (UTS) and University of Tokyo. Special thanks to Prof. S. Takizawa and Mitsubishi Rayon in providing the membrane module.

Special thanks must go to Professor Roger Ben Aim for his invaluable suggestions. I also would like to thank Dr. W. G. Shim for his assistance in mathematical modelling work and Mr. D. Hooper for his help in constructing the experimental unit.

This thesis would never have been completed without the great support from my beloved parents and friends.

Many thanks to the staffs and my office colleagues in Faculty of Engineering (UTS), Prof. H. Nguyen, Mr. L. Weber, Dr. P. Hagare, Ms R. Hamilton, Dr. T. Ramesh, Dr. D. S. Chaudhary, Van, Cuong, Vinh, Rong, Loan, Shingo, Paul, Shon, Nathaporn and Hugh.

Thank you very much to all of you!

TABLE OF CONTENTS

Acknowledgement	I
Table of contents	II
Nomenclature	IX
List of tables	XI
List of figures	XV
Abstract	XXIV

Chapter 1 Introduction	1-1
1.1 General	1-2
1.1.1 Wastewater reuse	1-2
1.1.2 Water reuse technologies	1-2
1.2 Objectives of the study	1-4
1.2.1 Crossflow microfiltration hybrid system	1-4
1.2.2 Submerged membrane adsorption hybrid system (SMAHS)	1-5
1.3 Scope of the study	1-5
1.3.1 Adsorption experiments	1-5
1.3.2 Flocculation experiments	1-6
1.3.3 Study on microfiltration hybrid system	1-6
1.3.4 Critical flux experiments	1-6
1.3.5 Study on submerged hollow fiber membrane-adsorption hybrid system (SMAHS)	1-7
1.3.6 Mathematical modelling of SMAHS	1-7
1.4 Organization of the report	1-7

Chapter 2 Literature Review	2-1
2.1 Introduction	2-2
2.2 The world's worsening water crisis	2-2
2.3 Wastewater reclamation and water reuse	2-2

2.4	Water reuse in Australia	2-5
2.5	Adsorption technology	2-8
2.5.1	General	2-8
2.5.2	Application of adsorption	2-10
2.5.3	Basic types of adsorbents	2-10
2.5.4	Activated carbon	2-11
2.5.4.1	General	2-11
2.5.4.2	Category and application of activated carbon	2-13
2.5.5	Powdered activated carbon (PAC)	2-14
2.5.6	Adsorption equilibria	2-16
2.5.6.1	Definition of adsorption Equilibria in liquid-phase	2-16
2.5.6.2	Langmuir adsorption isotherm	2-17
2.5.6.3	Freundlich adsorption isotherm	2-18
2.5.6.4	Langmuir-Freundlich (Sips) adsorption isotherm	2-19
2.5.6.5	Talu adsorption isotherm	2-20
2.5.7	Adsorption kinetics	2-21
2.5.7.1	General	2-21
2.5.7.2	Homogeneous surface diffusion model (HSDM)	2-23
2.6	Coagulation and flocculation technology	2-24
2.6.1	General	2-24
2.6.2	Flocculant chemicals	2-27
2.6.3	Ferric chloride	2-28
2.6.4	Flocculation-membrane system in water reuse	2-30
2.7	Membrane technology	2-31
2.7.1	General	2-31
2.7.2	Membrane processes and applications	2-31
2.8	Microfiltration	2-35
2.8.1	General	2-35
2.8.2	Microfiltration process	2-36
2.8.3	Membranes for microfiltration	2-38
2.8.4	Applications of microfiltration	2-38
2.9	Membrane fouling and critical flux	2-40
2.9.1	Membrane Fouling	2-40
2.9.2	Minimizing membrane fouling	2-42

2.9.3 Critical flux	2-44
2.9.3.1 General	2-44
2.9.3.2 Critical flux in crossflow microfiltration	2-46
2.10 Submerged Membrane Bioreactor Hybrid System	2-47
2.10.1 Organic pollutants in water and wastewater	2-48
2.10.1.1 General	2-48
2.10.1.2 The effects of organic pollutants	2-50
2.10.1.3 Organic pollutants removal	2-52
2.10.2 Biological wastewater treatment	2-53
2.10.2.1 Biological processes	2-53
2.10.2.2 Aerobic biological systems in wastewater treatment	2-55
2.10.2.3 Opportunities for membrane in biological wastewater treatment	2-56
2.10.3 Membrane bioreactor	2-59
2.10.3.1 General	2-59
2.10.3.2 Membrane separation in bioreactors	2-60
2.10.3.3 Membrane bioreactor as solids/liquid separation	2-62
2.10.4 Submerged membrane adsorption hybrid system	2-65

Chapter 3 Experimental investigation 3-1

3.1 Introduction	3-2
3.2 Wastewaters used in the experiments and their characteristics	3-2
3.2.1 Diluted synthetic wastewater	3-2
3.2.2 Synthetic secondary sewage effluent	3-2
3.2.3 Biological treated wastewater (sewage) effluent in Homebush	3-4
3.2.4 Biological treated wastewater (sewage) effluent in Gwangju	3-4
3.3 Adsorption equilibrium experiments	3-5
3.3.1 Methodology	3-5
3.3.2 Experimental conditions	3-6
3.4 Adsorption kinetics experiments	3-6
3.4.1 Methodology	3-7
3.4.2 Experimental conditions	3-8

3.5	Semi-pilot-scale on-site experiments	3-8
3.5.1	Methodology	3-8
3.5.2	Experimental conditions	3-11
3.6	Laboratory-scale critical flux experiments	3-13
3.6.1	Methodology	3-13
3.6.2	Experimental conditions	3-14
3.7	Experimental conditions optimization and modeling of submerged membrane adsorption hybrid system (SMAHS)	3-15
3.7.1	Methodology	3-17
3.7.2	Experimental conditions	3-20
3.8	Flocculation as pretreatment to SMAHS	3-20
3.8.1	Methodology	3-20
3.8.2	Experimental conditions	3-22
3.9	SMAHS experiment using Homebush biologically treated effluent and MWSD	3-23
3.10	Long term SMAHS experiments	3-23
3.10.1	Methodology	3-23
3.10.2	Experimental conditions	3-24
3.11	Measurement of molecular weight size distribution	3-25
3.10.1	Methodology	3-25
3.10.2	Determination of molecular weight size distribution	3-25

Chapter 4	Importance of pretreatment of crossflow microfiltration in tertiary wastewater treatment for reuse	4-1
------------------	---	------------

4.1	Introduction	4-2
4.2	Characterization of Homebush wastewater	4-4
4.3	Filtration quality	4-8
4.4	Effect of adsorption as pretreatment on the CFMF	4-9
4.4.1	Effect of PAC dose	4-9
4.4.2	Effect of initial filtration flux	4-10
4.4.3	Effect of backwash frequency	4-13
4.5	Effect of FMF as pretreatment on the CFMF	4-14

4.6 Flocculation and adsorption as pretreatment	4-16
4.7 Conclusions	4-18
 Chapter 5 Improving critical flux of CFMF through pretreatment	 5-1
5.1 Introduction	5-2
5.2 Critical flux experiments with synthetic wastewater	5-5
5.2.1 Performance of different pretreatments in terms of Total organic carbon (TOC) removal and modification on MW size distribution or organic matter	5-5
5.2.2 Short term critical flux experiments with different pretreatment methods	5-7
5.2.3 Long term critical flux experiments with different pretreatment methods	5-10
5.3 Effect of different crossflow velocities on critical flux of synthetic wastewater with and without pretreatment	5-13
5.3.1 Effect of crossflow velocity on critical flux of synthetic wastewater	5-14
5.3.2 Effect of crossflow velocity on critical flux for preflocculated synthetic wastewater	5-16
5.3.3 Effect of crossflow velocity on critical flux of synthetic wastewater after adsorption pretreatment	5-18
5.3.4 Effect of crossflow velocity on critical flux of synthetic wastewater after pretreatment of flocculation and adsorption	5-20
5.4 Critical flux experiments with biologically treated effluent in Homebush, Sydney	5-23
5.4.1 Performance of different pretreatments in terms of Total organic carbon (TOC) removal and modification on MW size distribution or organic matter	5-23
5.4.2 Short term critical flux experiments with different pretreatment methods	5-24
5.5 Critical flux experiments with biologically treated wastewater in Gwangju, South Korea	5-29
5.5.1 Critical flux experiments with different pretreatment methods	5-29
5.5.2 Effect of different pretreatments on organic matter removal and on MW size distribution	5-33

5.5.3 SEM investigation	5-37
5.6 Conclusions	5-40
 Chapter 6 Submerged membrane adsorption hybrid system: short term experiments and mathematical modelling	 6-1
6.1 Introduction	6-2
6.2 Mathematical modeling of membrane adsorption hybrid system	6-5
6.3 Adsorption equilibrium	6-10
6.4 Adsorption kinetics	6-10
6.5 Experimental conditions optimization and modeling of submerged membrane adsorption hybrid system (SMAHS)	6-11
6.5.1 Effect of preadsorption duration	6-11
6.5.2 Effect of aeration rate	6-14
6.5.3 Effect of backwash frequency	6-17
6.5.4 Effect of PAC dose	6-19
6.5.5 Effect of filtration flux	6-20
6.6 Flocculation as pretreatment to SMAHS	6-22
6.7 SMAHS experiment using Homebush biologically treated effluent and MWSD	6-27
6.8 Conclusions	6-30
 Chapter 7 Experimental investigation of submerged membrane adsorption hybrid system: long term experiments	 7-1
7.1 Introduction	7-2
7.2 Long term SMAHS experiments with diluted synthetic wastewater	7-4
7.3 The effect of PAC replacement of POPs removal in SMAHS	7-7
7.4 Long term Batch experiment on dry mass growth on PAC	7-9
7.5 Long term SMAHS experiments with synthetic secondary sewage effluent	7-11
7.5.1 Filtration flux of 24 L/m ² .h	7-11
7.5.2 Filtration flux of 12 L/m ² .h	7-15
7.6 Conclusions	7-18

Chapter 8	Conclusions and recommendations	8-1
8.1	Conclusions	8-2
8.1.1	Crossflow microfiltration hybrid system	8-2
8.1.2	Submerged membrane adsorption hybrid system (SMAHS)	8-4
8.1.2.1	Short term experiments	8-5
8.1.2.2	Long term experiments	8-6
8.2	Recommendations	8-7
References		R-1
Appendices		A-1
Appendix A	The critical flux of each (single) component in the synthetic wastewater	A-2
Appendix B	The stirred cell experiments	A-3
Appendix C	Publications	A-6

NOMENCLATURE

A_M	the surface area of the membrane (m^2)
b	adsorption affinity, a constant related to the heat of adsorption
C	concentration of the adsorbate in the solution (bulk phase concentration, mg/L)
C_b	the organic concentration in the bulk phase in the reactor (mg/L)
C_e	equilibrium concentration of the solute (mg/L)
C_e	effluent concentration
C_i	influent concentration
C_o	the organic concentration in the feeding tank (mg/L)
C_r	$Q\beta$ concentration in the bulk phase
C_r^∞	equilibrium concentration of $Q\beta$ in the bulk phase
C_s	the concentration of the external surface of PAC particles (mg/L)
D_e	the free liquid diffusivity of the solute
D_s	the surface diffusion coefficient (the rate of diffusion of the target compound along the surface of the carbon, m^2/s)
H	adsorption constant (Henry's Law)
k	the first order reaction coefficient
k_a	coefficient for adsorption onto PAC
k_m	coefficient for attachment to the membrane
k_d	coefficient for the bacterial decay, and
k_e	coefficient for inactivation due to the desorption of $Q\beta$ from PAC
K	constants characteristic of the system
K_f	the external mass transfer coefficient (m/s)
k_s	the solid mass transfer coefficient
M	the weight of PAC used (g)
MCC	the membrane correlation coefficient

n	parameter in the Sips equation
$1/n$	constants characteristic of the system
q	measured amount of organic matter adsorbed onto a unit amount of adsorbent (mg/g)
q^0	maximum adsorbed phase concentration (mg/g)
q_e	saturation amount of organic adsorbed (mg/g)
q_m	amount of solute adsorbed per unit weight of adsorbent required for monolayer capacity (mg/g)
q_t	the rate of change of surface concentration with time (t) at any radial distance (r) from the center of the activated carbon particle during adsorption (mg/g)
Q	the flow rate (m^3/s)
R	radius of carbon particle, L
T	temperature
V	the volume of the bulk solution in the reactor (m^3)
V_M	the volume of membrane (m^3)
W	PAC dose
$[(M/V) \cdot (dq/dt)]$	represents the adsorption of the organics onto PAC in suspension
$[(A_M/V_M) \cdot MCC \cdot C_b]$	describes the adsorption onto the PAC layer deposited onto membrane surface

Greek letters

ζ	parameter ($= \Psi(1 + K\Psi)$)
ψ	organic concentration spreading parameter
δ	the thickness of the diffusional sublayer
ρ_p	apparent density of the activated carbon (kg/m^3)

LIST OF TABLES

- Table 2.1 Water consumption for different activities in Australia (Australian Bureau of Statistics, 2000 and Sydney Water, 1999)
- Table 2.2 Annual water reuse from water utility STPs in Australia during 2001 and 2002 (Radcliffe, 2003a)
- Table 2.3 Recycled water use in State capital cities during 2001 and 2002 (Radcliffe, 2003b)
- Table 2.4 The quality of recycled water in Homebush Bay STP (Melbourne Water, 1999)
- Table 2.5 The practical applications of adsorption and related areas (Dąbrowski, 2001)
- Table 2.6 Basic types of common adsorbents (Dąbrowski, 2001)
- Table 2.7 Comparison of GAC and PAC Properties (AWWA, 1995)
- Table 2.8 Settling time for representative particles (Dharmappa and Hagare, 2000)
- Table 2.9 Common flocculation chemicals (AWWA, 1995)
- Table 2.10 Technically relevant main membrane processes in water treatment (Aptel and Buckley, 1996)
- Table 2.11 Application of membranes for treatment of wastewater allowing for the type of substance subject to treatment (Fane, 1996)
- Table 2.12 Comparison of various pressure-driven membrane processes (Mulder, 1996)
- Table 2.13 Milestones in the development of microfiltration (Baker, 2000)
- Table 2.14 Porosities and pore size distributions achieved by various preparation methods (Mulder, 1996)
- Table 2.15 Average foulant distribution worldwide (Van Hoof et al., 2002)
- Table 2.16 Methods of determining critical flux (Le Clech et al., 2003)
- Table 2.17 Composition of organic materials in wastewater (Levine et al., 1985)
- Table 2.18 Membrane configurations (Stephenson et al., 2000)
- Table 2.19 Membrane configurations (Stephenson et al., 2000)
- Table 2.20 Comparison of different configurations of MBR hybrid system (Seyfried et al., 1998)

Table 3.1 Constituents of the diluted synthetic wastewater

Table 3.2 Constituents of the synthetic wastewater

Table 3.3 Specific characteristics of biologically treated wastewater used over the experimental period (Homebush, Sydney)

Table 3.4 Characterization of biologically treated sewage effluent (Gwangju, South Korea)

Table 3.5 Characteristics of powdered activated carbon (PAC) used

Table 3.6 Adsorption equilibrium experimental conditions

Table 3.7 Adsorption kinetics experimental conditions

Table 3.8 Experimental conditions for filtrate quality experiments

Table 3.9 Experimental conditions for adsorption (PAC) as pretreatment to CFMF

Table 3.10 Experimental conditions for flocculation (FMF) as pretreatment to CFMF

Table 3.11 Experimental conditions for PAC and FMF as pretreatment to CFMF

Table 3.12 Experimental conditions for the short term and long term critical flux experiments using synthetic wastewater

Table 3.13 Experimental conditions for the short term critical flux experiments using biologically treated wastewater (sewage) effluent from Homebush WRT

Table 3.14 Experimental conditions to study the effect of different crossflow velocity on critical flux of synthetic wastewater

Table 3.15 Experimental conditions for the short term and long term critical flux using biologically treated effluent from Gwangju STP

Table 3.16 Characteristics of the hollow fiber membrane module

Table 3.17 Experimental conditions of short-term SMAHS experiments with synthetic secondary sewage effluent

Table 3.18 Experimental conditions of SF as pretreatment to SMAHS

Table 3.19 Experimental conditions of long term SMAHS experiments

Table 4.1 TOC removal efficiency of adsorption kinetics at different time (PAC dose = 1 g/L; initial TOC = 2.5519 mg/L; stirring speed = 110 rpm)

- Table 4.2 Effluent quality with different pretreatment (FMF velocity = 40 m/h; FeCl_3 = 50 mg/L; PAC dose = 1 g/L; PAC mixing time = 1-2 minutes; average influent TOC = 2.75 mg/L; experiment duration = 2 hours)
- Table 4.3 Comparison of filtration flux of adsorption-CFMF hybrid system at different dose of PAC (filtration flux = 200 L/m².h; membrane backwash frequency = 36 min)
- Table 4.4 The average value of TOC removal efficiency of adsorption-CFMF hybrid system operated at different initial filtration fluxes (PAC dose = 1 g/L; membrane backwash frequency = 36 min)
- Table 4.5 Comparison of average filtration flux of adsorption-CFMF hybrid system operated at different initial filtration fluxes (PAC dose = 1 g/L; membrane backwash frequency = 36 min)
- Table 4.6 Comparison of average filtration flux of adsorption-CFMF hybrid system for different backwash frequencies (PAC dose = 1 g/L; initial filtration flux = 250 L/m².h)
- Table 5.1 Effect of pretreatment on TOC removal of synthetic wastewater (Jar test)
- Table 5.2 Comparison of a long and short term critical flux of synthetic wastewater under different pretreatment conditions (synthetic wastewater; membrane pore size = 0.45 μm ; crossflow velocity = 0.15 m/s)
- Table 5.3 Average TOC removal efficiencies after different pre-treatments and CFMF (synthetic wastewater; membrane pore size = 0.45 μm ; crossflow velocity = 0.15 m/s)
- Table 5.4 The Reynolds number (Re) and shear stress values for different crossflow velocities
- Table 5.5 Effect of pretreatment on TOC removal of Homebush biologically treated effluent (Jar test)
- Table 5.6 The critical flux of biologically treated wastewater under different pretreatment conditions (crossflow velocity = 0.15 m/s; membrane pore size = 0.65 μm)
- Table 5.7 The critical flux of biologically treated wastewater under different pretreatment conditions (crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)
- Table 5.8 Average TOC removal efficiencies after different pre-treatments and CFMF (wastewater TOC = 6.8 mg/L; membrane pore size = 0.45 μm ; crossflow velocity = 0.15 m/s)
- Table 5.9 Effect of pretreatment on total organic carbon (TOC) removal of Gwangju biologically treated effluent (Jar test)

Table 6.1 The mass transfer coefficient in synthetic wastewater at different doses of PAC (initial TOC = 3.63 mg/L; stirring speed = 110 rpm)

Table 6.2 The modeling parameters of SMAHS at different PAC doses (filtration flux = 48 L/m².h; preadsorption = 1 hour; aeration rate = 16 L/min; backwash frequency = 1 hour; backwash duration = 1min; backwash rate = 120 L/m².h)

Table 6.3 Experimental conditions with different filtration flux during short-term experiments (PAC dose = 5 g/L; aeration rate = 16 L/min; preadsorption = 1 hour; backwash frequency = 1 hour; backwash duration = 1 min; backwash rate = 2.5 times of filtration flux)

Table 6.4 The modeling parameters of SMAHS at different filtration flux (PAC dose = 5 g/L; aeration rate = 16 L/min; preadsorption = 1 hour; backwash frequency = 1 hour; backwash duration = 1 min; backwash rate = 2.5 times of filtration flux)

Table 6.5 The average TOC removal efficiency at different operation configurations (filtration flux = 60 L/m².h; PAC dose = 2 g/L; FeCl₃ dose = 60 mg/L; flocculation velocity gradient = 30 s⁻¹; aeration rate = 12 L/min)

Table A1 The critical flux of single component (membrane pore size = 0.45 μm; crossflow velocity = 0.15 m/s)

LIST OF FIGURES

- Figure 2.1 Water quality changes during municipal uses of water in a time sequence (Asano, 2001)
- Figure 2.2 Schematic illustrating difference between dead-end and crossflow filtration (Ripperger, 1989)
- Figure 2.3 Schematic of various applications of MF: (a) MF only; (b) organics removal by MF pretreatment with coagulant or PAC; (c) MF as pretreatment to reverse osmosis; (d) MF as pretreatment to nanofiltration (Jacangelo and Buckley, 1996)
- Figure 2.4 Figure 2.4 Classification of organic substances present in natural water (Jegatheesan et al., 2002)
- Figure 2.5 Typical organic constituents in settled municipal wastewater (Levine et al., 1985)
- Figure 2.6 Evolution of water reclamation treatment processes (Côté et al., 1997)
- Figure 2.7 Schematics of MBR systems (Fane and Chang, 2002)
- Figure 3.1 Adsorption equilibrium experimental apparatus
- Figure 3.2 UV-Persulphate TOC analyzer (Dohrmann, phoenix 8000)
- Figure 3.3 Adsorption kinetics experimental apparatus
- Figure 3.4 CFMF unit
- Figure 3.5 Saturated floating medium flocculator and the medium (polystyrene beads)
- Figure 3.6 Schematic of flocculation-adsorption-microfiltration hybrid system
- Figure 3.7 The schematic diagram of the CFMF experimental set-up
- Figure 3.8 Laboratory-scale CFMF system
- Figure 3.9 Experimental set-up of SMAHS
- Figure 3.10 SMAHS system and hollow fiber membrane module
- Figure 3.11 Experimental set-up of SF-PAC-SMF hybrid system
- Figure 4.1 Influent variations over experimental period (turbidity (NTU); orthophosphate (PO_4^{-3} mg/ L); TOC (ppm C); suspended Solids (mg/ L))

Figure 4.2 MWSD of adsorption kinetics at different time (PAC dose = 1 g/L; initial TOC = 2.5519 mg/L; stirring speed = 110 rpm)

Figure 4.3 MWSD of adsorption as pretreatment (initial TOC = 2.5519 mg/L; stirring speed = 110 rpm; mixing time = 1 hour)

Figure 4.4 MWSD of flocculation as pretreatment (initial TOC = 2.5519 mg/L; rapid mixing = 1 min; slow mixing = 20 min)

Figure 4.5 MWSD of flocculation-adsorption as pretreatment (initial TOC = 2.5519 mg/L; adsorption stirring speed = 110 rpm; mixing time = 1 hour)

Figure 4.6 TOC removal efficiency of adsorption-CFMF system (influent TOC = 2.4–3 mg/L; initial filtration flux = 200 L/m².h; membrane backwash frequency = 36 min; PAC mixing speed = 125 rpm; PAC mixing time = 5 min)

Figure 4.7 TOC removal efficiency of adsorption-CFMF system (influent TOC = 2.4–3 mg/L; membrane backwash frequency = 36 min (backwash duration = 2 min); PAC mixing speed = 125 rpm)

Figure 4.8 TOC removal efficiency of adsorption-CFMF system (influent TOC = 2.4–3 mg/L; PAC dose = 1 g/L; membrane backwash frequency = 36 min; PAC mixing speed = 125 rpm; PAC mixing time = 5 min)

Figure 4.9 TOC removal efficiency of adsorption-CFMF system (influent TOC = 2.4–3 mg/L; PAC dose = 1 g/L; initial filtration flux = 250 L/m².h; PAC mixing speed = 125 rpm; PAC mixing time = 5 min)

Figure 4.10 Removal efficiency comparisons with and without flocculation as pretreatment (filtration flux: 220 L/m².h; membrane backwash frequency = 36 min (backwash duration 2 min); FMF velocity = 30 m/h; FeCl₃ dose = 50 mg/L; flocculator backwash frequency = 60 min (backwash duration 1 min))

Figure 4.11 Variation of filtration flux of CFMF with filtration time for different pretreatment (filtration flux = 220 L/m².h; membrane backwash frequency = 36 min; filtration velocity = 30 m/h; PAC = 1 g/L; FeCl₃ = 50 mg/L; flocculator backwash frequency = 60 min)

Figure 4.12 TOC removal at different stages of the flocculation-adsorption-microfiltration hybrid system (initial filtration flux = 220 L/m².h; membrane backwash frequency = 36 min; PAC = 1 g/L; FeCl₃ = 50 mg/L; FMF filtration velocity = 30 m/h; flocculator backwash frequency = 60 min)

Figure 4.13 TOC removal at different stages of the flocculation-adsorption-microfiltration hybrid system (initial filtration flux = 270 L/m².h; membrane backwash frequency = 36 min; PAC = 1 g/L; FeCl₃ = 50 mg/L; FMF filtration velocity = 30 m/h; flocculator backwash frequency = 60 min)

Figure 5.1 Comparison of flux profile with the flux below and above critical flux

Figure 5.2 MW distribution of synthetic wastewater with and without different pretreatments

Figure 5.3 CFMF experiments with constant filtration flux (synthetic wastewater; membrane pore size = $0.45\ \mu\text{m}$; crossflow velocity = $0.15\ \text{m/s}$)

Figure 5.4 CFMF experiments with constant filtration flux with flocculation as pretreatment (synthetic wastewater; membrane pore size = $0.45\ \mu\text{m}$; FeCl_3 dose = $68\ \text{mg/L}$; crossflow velocity = $0.15\ \text{m/s}$)

Figure 5.5 CFMF experiments with constant filtration flux with adsorption as pretreatment (synthetic wastewater; membrane pore size = $0.45\ \mu\text{m}$; PAC dose = $2\ \text{g/L}$; crossflow velocity = $0.15\ \text{m/s}$)

Figure 5.6 CFMF experiments with constant filtration flux of synthetic wastewater with flocculation-adsorption as pretreatment (synthetic wastewater; membrane pore size = $0.45\ \mu\text{m}$; PAC dose = $2\ \text{g/L}$; FeCl_3 = $68\ \text{mg/L}$; crossflow velocity = $0.15\ \text{m/s}$)

Figure 5.7 CFMF experiments with constant filtration flux (synthetic wastewater; membrane pore size = $0.45\ \mu\text{m}$; crossflow velocity = $0.15\ \text{m/s}$; filtration time at constant flux = 2 hour)

Figure 5.8 CFMF experiments with constant filtration flux with adsorption as pretreatment (synthetic wastewater; membrane pore size = $0.45\ \mu\text{m}$; PAC dose = $2\ \text{g/L}$; crossflow velocity = $0.15\ \text{m/s}$)

Figure 5.9 CFMF experiments with constant filtration flux with flocculation as pretreatment (synthetic wastewater; membrane pore size = $0.45\ \mu\text{m}$; FeCl_3 = $68\ \text{mg/L}$; crossflow velocity = $0.15\ \text{m/s}$)

Figure 5.10 CFMF experiments with constant filtration flux with flocculation-adsorption as pretreatment (synthetic wastewater; membrane pore size = $0.45\ \mu\text{m}$; PAC dose = $2\ \text{g/L}$; FeCl_3 = $68\ \text{mg/L}$; crossflow velocity = $0.15\ \text{m/s}$)

Figure 5.11 40 minutes constant permeate flux of synthetic wastewater at different crossflow velocity values (membrane pore size = $0.45\ \mu\text{m}$)

Figure 5.12 Effect of crossflow velocity on critical flux of preflocculated synthetic wastewater (membrane pore size = $0.45\ \mu\text{m}$)

Figure 5.13 Effect of crossflow velocity on critical flux of synthetic wastewater after adsorption as pretreatment (membrane pore size = $0.45\ \mu\text{m}$)

- Figure 5.14 Effect of crossflow velocity on critical flux of synthetic wastewater after flocculation and adsorption as pretreatment (membrane pore size = 0.45 μm)
- Figure 5.15 MW distribution of Homebush biologically treated effluent with and without different pretreatments
- Figure 5.16 CFMF experiments with constant filtration flux (Homebush biologically treated effluent; crossflow velocity = 0.15 m/s; membrane pore size = 0.65 μm)
- Figure 5.17 CFMF experiments with constant filtration flux after adsorption as pretreatment (Homebush biologically treated effluent; PAC dose = 2 g/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.65 μm)
- Figure 5.18 CFMF experiments with constant filtration flux after batch flocculation as pretreatment (Homebush biologically treated effluent; batch reactor mode; FeCl_3 = 50 mg/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.65 μm)
- Figure 5.19 CFMF experiments with constant filtration flux after FMF flocculation as pretreatment (Homebush biologically treated effluent; FMF mode, FeCl_3 = 50 mg/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.65 μm)
- Figure 5.20 Effect of flocculation (batch reactor) and adsorption as pretreatment on critical flux (Homebush biologically treated effluent; PAC = 2 g/L; FeCl_3 = 50 mg/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.65 μm)
- Figure 5.21 Effect of flocculation (FMF) and adsorption as pretreatment on critical flux (Homebush biologically treated effluent; PAC = 2 g/L; FeCl_3 = 50 mg/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.65 μm)
- Figure 5.22 CFMF experiments with constant filtration flux (Gwangju biologically treated effluent; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)
- Figure 5.23 CFMF experiments with constant filtration flux after batch flocculation as pretreatment (Gwangju biologically treated effluent; FeCl_3 = 120 mg/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)
- Figure 5.24 CFMF experiments with constant filtration flux after adsorption as pretreatment (Gwangju biologically treated effluent; PAC dose = 2 g/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)
- Figure 5.25 CFMF experiments with constant filtration flux after flocculation-adsorption as pretreatment (Gwangju biologically treated effluent; PAC = 2 g/L; FeCl_3 = 120 mg/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)

Figure 5.26 Long term CFMF experiments with constant filtration flux with flocculation-adsorption as pretreatment (Gwangju biologically treated effluent; PAC dose = 2 g/L; FeCl_3 = 120 mg/L; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)

Figure 5.27 MW distribution of Gwangju biologically treated effluent with and without different pretreatments

Figure 5.28 MW distribution of biologically treated effluent without any pretreatment (Gwangju wastewater water; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)

Figure 5.29 MW distribution of biologically treated effluent after flocculation (Gwangju wastewater water; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)

Figure 5.30 MW distribution of biologically treated effluent after adsorption (Gwangju wastewater water; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)

Figure 5.31 SEM of new membrane and membrane after critical flux with different pretreatment (Gwangju wastewater water; crossflow velocity = 0.15 m/s; membrane pore size = 0.45 μm)

Figure 6.1 Adsorption isotherm of synthetic wastewater (initial TOC = 3.87 mg/L)

Figure 6.2 Adsorption kinetics of synthetic wastewater at different doses of PAC (initial TOC = 3.63 mg/L; stirring speed 110 rpm; C = effluent TOC concentration, mg/L and C_0 = influent TOC concentration, mg/L)

Figure 6.3 TOC removal efficiency of effluent at different durations of preadsorption (filtration flux = 48 $\text{L}/\text{m}^2\cdot\text{h}$; PAC dose = 5 g/L; aeration rate = 8 L/min)

Figure 6.4 TMP profile at different durations of preadsorption (filtration flux = 48 $\text{L}/\text{m}^2\cdot\text{h}$; PAC dose = 5 g/L; aeration rate = 8 L/min)

Figure 6.5 TOC removal efficiency of PAC in PAC-MF reactor at different durations of preadsorption (filtration flux = 48 $\text{L}/\text{m}^2\cdot\text{h}$; PAC dose = 5 g/L; aeration rate = 8 L/min)

Figure 6.6 Model prediction of effluent concentration at different aeration rates (filtration flux = 48 $\text{L}/\text{m}^2\cdot\text{h}$; PAC = 5 g/L; preadsorption = 1 hour; $D_s = 4.709 \times 10^{-15}$)

Figure 6.7 Variation of the model parameters of SMAHS at different aeration rates (filtration flux = 48 $\text{L}/\text{m}^2\cdot\text{h}$; PAC = 5 g/L; Preadsorption = 1 hour; $D_s = 4.709 \times 10^{-15}$)

Figure 6.8 TOC removal efficiency at different aeration rates (filtration flux = 48 $\text{L}/\text{m}^2\cdot\text{h}$; PAC dose = 5 g/L; preadsorption = 1 hour)

Figure 6.9 TOC removal efficiency at different backwash frequencies (filtration flux = 48 L/m².h; PAC dose = 2 g/L; preadsorption = 1 hour; aeration rate = 16 L/min)

Figure 6.10 TMP profile at different backwash frequencies (filtration flux = 48 L/m².h; PAC dose = 2 g/L; preadsorption = 1 hour; aeration rate = 16 L/min)

Figure 6.11 Model prediction of organic removal at different PAC doses (filtration flux = 48 L/m².h; preadsorption = 1 hour; aeration rate = 16 L/min; backwash frequency = 1 hour; backwash duration = 1min; backwash rate = 120 L/m².h; $D_s = 4.709 \times 10^{-15}$; C = effluent TOC concentration, mg/L and C_o = influent TOC concentration, mg/L)

Figure 6.12 Effect of PAC dose on the TMP development (filtration flux = 48 L/m².h; preadsorption = 1 hour; aeration rate = 16 L/min; backwash frequency = 1 hour; backwash duration = 1 min; backwash rate = 120 L/m².h)

Figure 6.13 Model prediction of organic removal at different filtration flux (PAC dose = 5 g/L; preadsorption = 1 hour; aeration rate = 16 L/min; backwash frequency = 1 hour; backwash duration = 1min; backwash rate = 2.5 times of filtration flux; $D_s = 4.709 \times 10^{-15}$; C = effluent TOC concentration, mg/L and C_o = influent TOC concentration, mg/L)

Figure 6.14 Effect of different filtration flux on the TMP development (PAC dose = 5 g/L; preadsorption = 1 hour; aeration rate = 16 L/min; backwash frequency = 1 hour; backwash duration = 1min; backwash rate = 2.5 times of filtration flux)

Figure 6.15 TMP profile of SMF and PAC-SMF versus operation time (filtration flux = 60 L/m².h; aeration rate = 12 L/min)

Figure 6.16 TMP profile of PAC-SMF at two different doses of PAC (filtration flux = 60 L/m².h; aeration rate = 12 L/min)

Figure 6.17 TMP profile of SF-SMF with flocs settling and non-settling (filtration flux = 60 L/m².h; FeCl₃ dose = 60 mg/L; flocculation velocity gradient = 30 s⁻¹; aeration rate = 12 L/min)

Figure 6.18 TMP profile of SF-PAC-SMF with flocs settling and non-settling (filtration flux = 60 L/m².h; PAC dose = 2 g/L; FeCl₃ dose = 60 mg/L; flocculation velocity gradient = 30 s⁻¹; aeration rate = 12 L/min)

Figure 6.19 TOC removal efficiency at different operation configurations (filtration flux = 60 L/m².h; PAC dose = 2 g/L; FeCl₃ dose = 60 mg/L; flocculation velocity gradient = 30 s⁻¹; aeration rate = 12 L/min)

Figure 6.20 Comparison of TOC removal efficiency of SMAHS in case of using synthetic wastewater and biologically treated effluent (PAC dose = 5 g/L; preadsorption = 1 hour; aeration rate = 16 L/min; backwash frequency = 1 hour 1 min; backwash rate = 2.5 times of filtration rate)

Figure 6.21 Comparison of TMP development of SMAHS in case of using synthetic wastewater and biologically treated effluent (PAC dose = 5 g/L; preadsorption = 1 hour; aeration rate = 16 L/min; backwash frequency = 1 hour 1 min; backwash rate = 2.5 times of filtration rate)

Figure 6.22 MWSD of the biologically treated effluent in the SMAHS (operation duration = hour)

Figure 6.23 MWSD of the biologically treated effluent in the SMAHS (operation duration 5 = hour)

Figure 6.24 MWSD of the biologically treated effluent in the SMAHS (operation duration = 10 hour)

Figure 7.1 TOC removal efficiency of the submerged membrane hybrid system without backwash (synthetic wastewater; influent TOC = 2-3.5 mg/L; PAC dose = 1 g/L; filtration flux = $0.288 \text{ m}^3/\text{m}^2\text{d}$; aeration rate = 8 L/min)

Figure 7.2 TMP development of the submerged membrane hybrid system without backwash (synthetic wastewater; influent TOC = 2-3.5 mg/L; PAC dose = 1 g/L; filtration flux = $0.288 \text{ m}^3/\text{m}^2\text{d}$; aeration rate = 8 L/min)

Figure 7.3 TOC removal efficiency of the submerged membrane hybrid system with daily-backwash (synthetic wastewater; influent TOC = 2-3.5 mg/L, PAC dose = 5 g/L; filtration flux = $0.288 \text{ m}^3/\text{m}^2\text{d}$, backwash frequency = 1 day; backwash rate = $87 \text{ m}^3/\text{m}^2\text{d}$; backwash duration = 2 minutes; aeration rate = 8 L/min)

Figure 7.4 TMP development of the submerged membrane hybrid system with daily-backwash (synthetic wastewater; influent TOC = 2-3.5 mg/L, PAC dose = 5 g/L; filtration flux = $0.288 \text{ m}^3/\text{m}^2\text{d}$, backwash frequency = 1 day; backwash rate = $87 \text{ m}^3/\text{m}^2\text{d}$; backwash duration = 2 minutes; aeration rate = 8 L/min)

Figure 7.5 TOC removal efficiency versus operation time (PAC dose = 5 g/L; filtration rate = $12 \text{ L}/\text{m}^2\text{.h}$; aeration rate = 12 L/min; HRT = 0.56 day; backwash frequency = 2 times/day; backwash duration = 2 minutes; backwash rate = $30 \text{ L}/\text{m}^2\text{.h}$)

Figure 7.6 Variation of TOC of influent, PAC-MF reactor and effluent (PAC dose = 5 g/L; filtration rate = $12 \text{ L}/\text{m}^2\text{.h}$; aeration rate = 12 L/min; HRT = 0.56 day; backwash frequency = 2 times/day; backwash duration = 2 minutes; backwash rate = $30 \text{ L}/\text{m}^2\text{.h}$)

Figure 7.7 TOC removal efficiency of dry mass growth experiment (PAC dose = 5 g/L; aeration rate = 16 L/min)

Figure 7.8 Dry mass growth during experiment (PAC dose = 5 g/L; aeration rate = 16 L/min)

Figure 7.9 Molecular weight size distribution of dry mass growth experiment (PAC dose = 5 g/L; aeration rate = 16 L/min)

Figure 7.10 TOC removal efficiency of SMAHS experiment at filtration flux of 24 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.21 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.11 TMP development of SMAHS experiment at filtration flux of 24 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.21 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.12 Dry mass growth of SMAHS experiment at filtration flux of 24 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.21 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.13 DO concentration variation of SMAHS experiment at filtration flux of 24 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.21 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.14 The 14th day MWSD of SMAHS experiment at filtration flux of 24 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.21 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.15 TOC removal efficiency of SMAHS experiment at filtration flux of 12 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.42 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.16 TMP development of SMAHS experiment at filtration flux of 24 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.42 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.17 DO concentration variation of the wastewater taken at different periods at filtration flux of 12 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.42 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.18 The actual oxygen consumption rate of SMAHS experiment at filtration flux of 12 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.42 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure 7.19 Dry mass growth of SMAHS experiment at filtration flux of 12 L/m².h (PAC dose = 5 g/L; aeration rate = 16 L/min; HRT = 0.42 day; backwash frequency = 1 hour; backwash duration = 1 minutes; backwash rate = 30 L/m².h)

Figure B1 The experimental set-up of stirred cell

Figure B2 The permeate flux decline of synthetic wastewater with and without adsorption as pretreatment (influent TOC = 2.8176 mg/L; TMP = 150 kPa)

Figure B3 The permeate flux decline of Homebush wastewater with and without flocculation as pretreatment (initial wastewater TOC = 3.7744 mg/L; TMP = 150 kPa; FMF velocity = 30 m/h; FeCl₃ dose = 50 mg/L; flocculator backwash frequency = 60 min (backwash duration 1 min))

Figure B4 The permeate flux decline of Homebush wastewater with and without adsorption as pretreatment (initial wastewater TOC = 3.7744 mg/L; TMP = 150 kPa)

Figure B5 The permeate flux decline of flocculated Homebush wastewater with and without adsorption as pretreatment (initial wastewater TOC = 3.7744 mg/L; TMP = 150 kPa; FMF velocity = 30 m/h; FeCl₃ dose = 50 mg/L; flocculator backwash frequency = 60 min (backwash duration 1 min))

ABSTRACT

Generally, the conventional wastewater treatment cannot remove all the effluent organic matter (EfOM) such as synthetic organic chemicals and natural organic matter etc. As a result, the biologically treated effluent from sewage treatment plant needs to undergo further advanced treatment processes. To obtain water of recyclable quality, initially physico-chemical processes such as flocculation, sedimentation, filtration and adsorption were normally used. However, with advanced technologies and ever increasing stringent water quality criteria, membrane processes are becoming more attractive in water reuse.

Among different membrane processes, although microfiltration (MF) can be operated economically, it alone cannot remove organic matter. If MF is combined with an enhanced flocculation or/and adsorption, it will be able to reduce superior level of organic contaminants. The aims of this study are: (i) improving the dissolved organic removal and reduce membrane fouling of two membrane hybrid systems (crossflow microfiltration (CFMF) and submerged membrane adsorption hybrid system (SMAHS)) using different pretreatment methods (flocculation, adsorption and flocculation-adsorption); (ii) investigating the critical flux of a laboratory-scale CFMF with and without different pretreatments.

The incorporation of powdered activated carbon (PAC) as pretreatment to CFMF resulted in high TOC removal efficiency (more than 80%) when the PAC-CFMF system was operated at a relatively high filtration flux of $250 \text{ L/m}^2\cdot\text{h}$. The incorporation of flocculation and PAC as pretreatments to CFMF process resulted in a very high TOC removal efficiency (99.7%) and a stable filtration flux during 5-hour filter run (less than 12% flux decline), when the hybrid system was operated at a higher filtration flux ($270 \text{ L/m}^2\cdot\text{h}$).

Application of membrane processes requires lower investment and operating costs. One of the ways is to operate system at a constant filtration flux below the critical flux. With both flocculation and adsorption as pretreatment to CFMF, the critical flux of biologically treated effluent increased dramatically (5-7 times increase).

The preadsorption, PAC dose, aeration rate and filtration flux had effects both on organic matter removal efficiency and TMP development. The preadsorption of 1 hour prior to the membrane operation was important in mitigating the membrane fouling. The suitable aeration rate, filtration flux and initial PAC dosing were 16 L/min, $< 24 \text{ L/m}^2\cdot\text{h}$ and 5 g/L respectively for the wastewater used in this study. The long term SMAHS experiments conducted with regular PAC replacement indicated that the PAC replacement in PAC-MF reactor could stimulate both biological activity and adsorption, as well as optimize the operation of the hybrid system.